

Title: Development of Multi-Task Catalysts for Removal of NO_x, and Toxic Organic Compounds during Coal Combustion (DE-FG26-97FT97274)

Authors: Panagiotis G. Smirniotis, Assistant Professor
e-mail: Panagiotis.Smirniotis@UC.EDU
phone (513) 556-1474; fax (513) 556-3473
Chemical Engineering Department, University of Cincinnati
P.O. Box 210171
Cincinnati, OH 45221-0171

Robert G. Jenkins, Professor and Interim Dean of the Engineering College
e-mail: Robert.Jenkins@UC.EDU
phone (513) 556-2933; fax (513) 556-3626
Chemical Engineering Department, University of Cincinnati
P.O. Box 210018
Cincinnati, OH 45221-0018

ABSTRACT

The work performed during the first six months on this project was a preliminary screening for effectiveness of selected oxide-based catalysts for the SCR of NO_x. The catalytic tests were performed with a "synthetic" flue gas containing NO, O₂, and NH₃ (reductant) with a range of mixed oxide catalysts. Experiments with organics simulating typical VOCs which coexist in the flue gas, such as benzene and chlorobenzene will start during the summer 1998. The catalysts employed in this study include mixed oxide composite powders (combinations of TiO₂ (primary oxide), Cr₂O₃, ZrO₂, WO₃, SiO₂, and Al₂O₃) loaded with varying amounts of V₂O₅. The mixed oxide supports were prepared from inorganic precursors by sol-gel co-precipitation. Vanadia was added to the supports using wet impregnation methods. Commercial titania was also impregnated with vanadia and tested under the same conditions for comparison purposes.

The W/TiO₂-based catalysts yielded excellent conversion and selectivity in the SCR of NO_x. We specifically found that these catalysts exhibited complete NO_x conversion and more than 95 % N₂ selectivity over a wide reaction temperature range (250 C to 375 C). The corresponding range for optimum performance over the commercial catalyst, which was worse than the W/TiO₂-based catalysts, was from 250 C to 310 C. This is a very important characteristic because higher reaction temperatures can be used to more efficiently oxidize the organics. It should be noted that Cr and Zr were added to TiO₂ since they favor oxidation reactions and therefore can potentially oxidize VOCs. It was found that for an organic free stream both dopants have a negative effect because they decrease the DeNO_x capabilities of the catalyst. Commercial titania impregnated with WO₃ and loaded with vanadia demonstrated similar N₂ selectivity trends but significantly lower NO conversion at any temperature in comparison with the sol-gel W/TiO₂ supports. TiO₂/SiO₂/Al₂O₃ composites were prepared and tested under the same conditions. The NO conversion as well as N₂ selectivity was slightly lower than the W/TiO₂-based catalysts at any temperature. Increasing of the vanadia content of the catalysts has a positive effect on the NO conversion but reduces the N₂ selectivity.

Anatase was the only titania phase identified in our catalysts from XRD spectra. The percent of titania crystallinity of our supports was relatively low. Although previous reports stated that high titania crystallinity was necessary for good SCR performance, we found that this was not the case over our mixed oxide catalysts. It is believed that the high support BET area ($130 \text{ m}^2/\text{g}$) and increased Brønsted to Lewis acid ratio of the W/TiO₂-based catalysts both contribute to overcome the disadvantage of low crystallinity. Supporting evidence of the increased acidity comes from FT-IR and stepwise programmed desorption of NH₃ experiments. These experiments show a significant increase of Brønsted acidity (band at 1426 cm^{-1}) for the tungsten rich catalysts. Lewis acidity (band at 1605 cm^{-1}) remained at the same levels with pure titania-supported catalysts. Both techniques indicate that the Lewis and Brønsted sites are of comparable strength and disappear at temperatures higher than 450 C. It was also concluded that the method employed for the loading of W is very crucial (Economidis et al., 1998; Economidis, 1998) for the synthesis of a successful catalyst. From preliminary results with cofed benzene over the V₂O₅/W-TiO₂ catalysts, substantial oxidation of the organic takes place even though the oxygen feed concentration was well below the values reported by other researchers. When the oxygen concentration was increased to 2%, almost complete oxidation of benzene was achieved. This is a strong indication that these systems will probably be effective for DeNO_x reactions in the presence of organics.

The bench-scale mini-combustor, that will be utilized for the production of flue gas from coal is being constructed. Some more electronic work (wiring, installment of the data acquisition system) needs to be done to complete this task. It is expected that the first experiments with the combustor will start early this summer.

REFERENCES:

- N. Economidis, R. F. Coil, and P. G. Smirniotis, Catalytic Performance of Al₂O₃/SiO₂/TiO₂ loaded with V₂O₅ for the Selective Catalytic Reduction of NO_x with Ammonia, *Catalysis Today*, Vol. 40, 27-37, 1998.
- N. Economidis, Master Thesis, University of Cincinnati, 1998.

List of published Journal Articles, Completed Presentations, and Students Receiving Support from the Grant

Articles/Presentations. The inception of the grant was October 1997. The first manuscript is in preparation from the experimental work of Mr. Donovan Peña who joined the group in November 1997.

Students Receiving Support from the Grant.

Graduate Students: Mr. Donovan Peña, Mr. Wenmin Zhang, and Ms. Elizabeth Allen.

Undergraduate Students: Mr. Benjamin Caundill, Mr. Matt Gherinng, and Mr. Brad Hedden.